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# Bioaccumulation of DDT and PCB in Tissues of Marine Fishes

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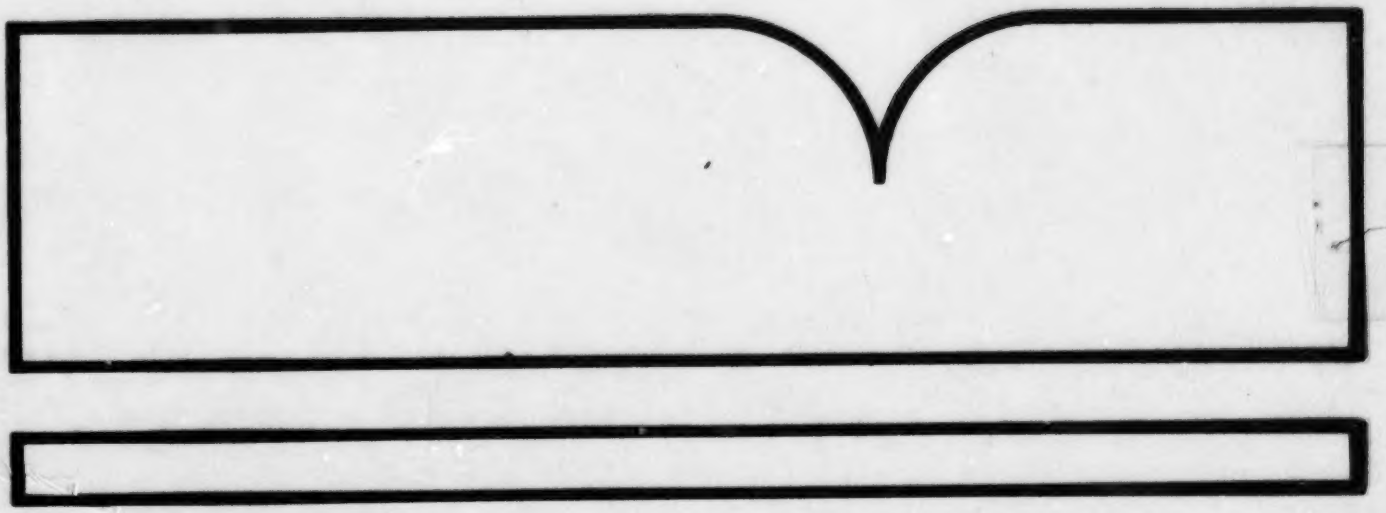

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The proportions of the DDT metabolites found suggest that this pesticide had been accumulating in the 18-20 year period of maturation of the female and was passed on to the first brood of young. In contrast to the findings of other investigations, there was no fixed relationship in the relative magnitude of DDT and PCB residues when both compounds were present in a sample.

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# Bioaccumulation of DDT and PCB in Tissues of Marine Fishes

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**ABSTRACT:** Fishes of commercial importance were monitored in New England coastal waters in 1974 to determine whether synthetic organic residues in the fish were large enough to affect the utilization of such fish as food by man or to interfere with their ability to reproduce. About 700 fish of 20 species were pooled in samples of five to ten, and the livers were analyzed. Several species, including the spiny dogfish, contained residues of dichlorodiphenyltrichloroethane (DDT) and its metabolites and of polychlorinated biphenyl (PCB) compounds in the 1 to 10 μg/g (ppm) range. More detailed studies of the dogfish in 1975 demonstrated the transfer of these compounds from the parent fish to the ovarian egg and the mature fetus.

The proportions of the DDT metaboljtes found suggest that this pesticide had been accumulating in the 18 to 20-year period of maturation of the female and was passed on to the first brood of young. In contrast to the findings of other investigations, there was no fixed relationship in the relative magnitude of DDT and PCB residues when both compounds were present in a sample.

**KEY WORDS:** DDT, PCB, bioaccumulation, monitoring, aquatic toxicology

Domestic use of dichlorodiphenyltrichloroethane (DDT) in the United States reached its height in 1959; by 1966 its use was sharply curtailed, and, by 1969, it was banned from a majority of formerly widespread environmental uses. Its environmental persistence has been amply demonstrated in an almost infinite variety of biotic samples, but there has been no systematic monitoring of marine fishes comparable to the national programs for monitoring molluscs [*1*][3] and estuarine fishes [*2*].

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[3] The italic numbers in brackets refer to the list of references appended to this paper.

The National Marine Fisheries Service (NMFS) conducts annual surveys of bottom fish resources on important commercial fishing grounds. In 1974, the NMFS arranged to have representative surplus fish from these surveys analyzed by chemists from the Environmental Protection Agency (EPA) to determine whether the residues of pesticides or polychlorinated biphenyl (PCB) compounds present were of such magnitude as to endanger the fish or make them unfit for human food. Fish from the New England coast are discussed in this report.

## Methods

Twenty species of fish, 14 of them commercially important, were selected from trawls at about 80 stations irregularly scattered in an area from 5 to 200 miles off the coast and extending from New Jersey to Nova Scotia. The fish were frozen immediately in special aluminum-lined kraft bags, and transported to the NMFS Center for refrigerated storage (Table 1). The liver tissues were analyzed because the residues of halogenated organics

TABLE 1—*List of fish species sampled.*

| Common Name | Scientific Name |
|---|---|
| Alewife | *Alosa pseudoharengus* |
| American plaice | *Hippoglossoides platessoides* |
| Butterfish | *Peprilus triacanthus* |
| Cod | *Gadus morhua* |
| Fourspot flounder | *Paralichthys oblongus* |
| Goosefish | *Lophius americanus* |
| Haddock | *Melanogrammus aeglefinus* |
| Little skate | *Raja erinacea* |
| Longhorn sculpin | *Myoxocephalus octodecemspinosus* |
| Mackerel | *Scomber scombrus* |
| Northern searobin | *Prionotus carolinus* |
| Ocean pout | *Macrozoarces americanus* |
| Pollock | *Theragra chalcogramma* |
| Redfish | *Sebastes marinus* |
| Red hake | *Urophycis chuss* |
| Scup | *Stenotomus chrysops* |
| Sea herring | *Clupea harengus harengus* |
| Silver hake | *Merluccius bilinearis* |
| Spiny dogfish | *Squalus acanthias* |
| Yellowtail flounder | *Limanda ferruginea* |

are typically largest in this organ because of its high lipid content. Replicate pooled samples of 5 to 10 fish, a total of 668, were analyzed by gas chromatographic techniques, as described elsewhere [2]. The residue data are reported in micrograms per gram (wet weight) of tissue. All the samples contained measureable amounts of DDT, or one of its metabolites, and PCBs. The PCBs were quantified with Aroclor 1254 as the standard.

**Results and Discussion**

Total DDT and PCB levels in the livers of New England Coastal Zone fishes ranged from 0.02 to 4.76 μg/g and from 0.01 to 2.47 μg/g, respectively (Table 2). There are some difficulties in the interpretation of these data. Of particular importance is the lack of uniformity in the size and age of the fish. A gradual increase in pesticide residues with time has been clearly shown in several species of marine and freshwater fishes, and this factor could affect these data [*3,4*].

A second difficulty resulted from the pooling of fish collected at stations some distance apart in order to have an adequate number of individuals per sample. In some cases, for example, there were characteristic residue differences in samples of the same species and same size taken at different geographical locations. These differences are lost when pooling either the fish or the data.

The levels of ΣDDT (the sum of the residues of DDT and its metabolites) and of PCB compounds in the livers of Group I fish may be considered the current background levels in many fish species in estuaries of the eastern United States [*2,5*]. The residues in Group II fish are about an order of magnitude larger. They still are of little concern because, in most instances in which liver and muscle tissue from the same fish were analyzed, the liver residues of 1 to 2 μg/g were not accompanied by detectable residues in the edible muscle. However, the residues of ΣDDT in the spiny dogfish were high enough to suggest some special bioaccumulation conditions.

At one station about 50 miles east of Cape Cod, the DDT residue in sculpins was 25 percent that of the PCBs. At a second station 100 miles farther east, but at the same bottom depths, the DDT residues were twice as much as the PCB residues in sculpins of similar size. Although it has been stated that PCB residues are always at least ten times higher than DDT residues in marine biota from this general area, this was clearly not the case in 19 of the 20 species we sampled [*6*].

Additional information may be gained from these analyses when the total DDT residues is recorded in terms of the percentage distribution of the metabolites dichlorodiphenyldichloroethylene (DDE) and tetrachlorodiphenylethane (TDE), and the parent compound (Table 3). The Group I data have a percentage distribution of the metabolites that is fairly typical for recent biological samples, and the predominance of DDE indicates its resistance to metabolic decay. The somewhat lower levels of DDE in Group II are a little unexpected, whereas the high proportion of the parent compound, DDT, in dogfish livers is surprising. The identity of this component was confirmed by mass spectography. The predominance of the parent compound in tissue residues is more characteristic of recent contamination and is not expected in fish collected 100 miles offshore about a decade after the use of DDT had virtually terminated.

TABLE 2—*Average residue values of ΣDDT and PCB in North Atlantic marine fish livers (μg/g wet weight).*[a]

| Group I | | | | Group II | | | |
|---|---|---|---|---|---|---|---|
| Common Name | No. of Fish | ΣDDT | PCB | Common Name | No. of Fish | ΣDDT | PCB |
| Yellowtail flounder | 40 | 0.02 | 0.13 | Northern searobin | 24 | 0.13 | 0.39 |
| Butterfish | 40 | 0.02 | 0.54 | Mackerel | 30 | 0.19 | 0.33 |
| American plaice | 20 | 0.05 | 0.20 | Haddock | 30 | 0.32 | 0.72 |
| Fourspot flounder | 38 | 0.05 | 0.28 | Red hake | 30 | 0.35 | 0.96 |
| Ocean pout | 50 | 0.07 | 0.31 | Redfish | 24 | 0.35 | 0.34 |
| Scup | 40 | 0.07 | 0.35 | Little skate | 20 | 0.37 | 1.26 |
| Alewife | 40 | 0.09 | 0.27 | Silver hake | 40 | 0.44 | 1.01 |
| Sea herring | 40 | 0.09 | 0.13 | Cod | 20 | 0.70 | 1.12 |
| Longhorn sculpin | 54 | 0.10 | 0.10 | Pollock | 28 | 1.06 | 2.27 |
| | | | | Goosefish | 34 | 1.35 | 2.76 |
| $\bar{X}$ | | 0.06 | 0.26 | $\bar{X}$ | | 0.55 | 1.12 |
| $\sigma$ | | 0.03 | 0.14 | $\sigma$ | | 0.44 | 0.82 |
| Spiny dogfish | 20 | 4.76 | 2.47 | | | | |

[a] Species are grouped on the basis of having more or less than 0.11 μg/g of DDT. The values are the means of two or more replicate samples of equal numbers.

TABLE 3—*Average percentage distribution of DDT metabolites in marine fish livers.*[a]

| | DDT | TDE | DDE |
|---|---|---|---|
| Group I | 11 | 19 | 70 |
| Group II | 30 | 25 | 45 |
| Spiny dogfish | 58 | 13 | 29 |

[a] Groups I and II are the same as those shown in Table 2.

The kinetics of DDT in the estuarine food web are well illustrated by data from analyses of fish monitored in North Carolina during the past few years (Table 4). In that program, only young-of-the-year fish were sampled; consequently, any DDT residues detected had accumulated in less than the previous twelve-month period [*2*]. These data demonstrate the gradual disappearance of DDT from the environment and the estuarine food web. It is apparent that in the recycling of DDT, the parent compound was converted to the more persistent metabolites. The magnitude of the individual ΣDDT residues did not change significantly, but the number of positive samples (> 9 μg/kg) declined during the monitoring period. The data also indicate, by the absence of the parent compound, that there was no significant reintroduction of DDT into North Carolina estuaries in recent years.

By extrapolation from these data, we may infer that Group I fish were mostly less than four years old, and Group II fish included more older individuals. This is in general agreement with our knowledge of the age of fish caught in the commercial fishery.

The high percentage of DDT in dogfish livers remained unexplained since it seemed unlikely that they had been exposed recently to a primary source of DDT pollution. This situation appeared to be peculiar to dogfish from the New England Coastal Zone since these investigators had monitored other populations of dogfish in North Carolina and Washington and found typically low residues, consisting primarily of DDE.

TABLE 4—*Distribution of metabolites in ΣDDT residues in estuarine yearling fish in North Carolina.*[a]

| Year | No. of Samples | Samples with ΣDDT, % | DDT, % | TDE, % | DDE, % | Geographic $\overline{X}$ of ΣDDT in Positive Samples, μg/g |
|---|---|---|---|---|---|---|
| 1972 | 30 | 97 | 31 | 25 | 44 | 0.04 |
| 1973 | 80 | 43 | 13 | 29 | 58 | 0.04 |
| 1974 | 66 | 38 | 6 | 31 | 63 | 0.04 |
| 1975 | 40 | 45 | 0 | 22 | 78 | 0.02 |
| 1976 | 30 | 18 | 0 | 18 | 82 | 0.03 |

[a] Samples consisted of 50 fish that were analyzed in replicate pools of 25 each.

The authors decided to investigate the problem further, and, in the spring of 1975, collected uniformly small fish of four of the species that had had the highest DDT residues the year before. These fish were trawled in the same general area as the 1974 collections. With the exception of the dogfish, the livers of the smaller fish contained the anticipated low residue levels and corroborated the opinion that the larger DDT residues of Group II fish represented the accumulation of several years (Table 5).

The dogfish enigma was unsolved. The livers of juvenile dogfish from coastal New England and New Jersey contained DDT residues as high as and higher than those found in samples from older sharks collected a year earlier. The ΣDDT residues were, again, as much as three times larger than the residues of PCB. These juvenile dogfish were as small as neonates (25 to 32 cm), and it was noted that all had empty digestive tracts. It seemed likely that they had acquired the residues before birth.

The NMFS helped us complete this investigation by catching 14 gravid sharks in the fall of 1975 at eight stations on the same general fishing grounds (Table 6). The oviducts of these dogfish contained about 100 developmental stages, which ranged from mature eggs to mature fetuses and included embryos. These embryonic tissues were removed, and similar developmental stages were analyzed in pooled samples. To the extent possible, samples were made up of tissues from the same mother and contained from two to twelve separate tissues.

The results of these analyses confirmed the hypothesis that the DDT and PCB residues are acquired from the mother during embryonic development (Table 7). The similarity of the residue concentrations in the yolk and the

TABLE 5—*Residues of DDT, its metabolites, and PCB in pooled livers of young marine fish.*[a]

| Common name | No. of Fish | DDT | TDE | DDE | ΣDDT | PCB |
|---|---|---|---|---|---|---|
| Goosefish | 10 | ... | ... | 0.13 | 0.13 | 0.25 |
| | 9 | ... | ... | 0.04 | 0.04 | 0.63 |
| | 7 | ... | ... | 0.03 | 0.03 | 0.39 |
| Little skate | 11 | ... | ... | 0.35 | 0.35 | ... |
| | 10 | ... | ... | 0.42 | 0.42 | ... |
| Pollock | 10 | ... | ... | 0.19 | 0.19 | ... |
| | 12 | ... | 0.33 | 0.31 | 0.64 | 0.21 |
| Dogfish | 10 | 1.12 | 0.44 | 1.38 | 1.94 | 2.80 |
| | 10 | 2.12 | 0.88 | 1.67 | 4.67 | 2.03 |
| | 12 | 2.23 | 0.59 | 2.46 | 5.28 | 4.66 |
| | 10 | 3.64 | 1.57 | 4.00 | 9.21 | 3.74 |
| | 10 | 3.88 | 1.60 | 4.00 | 9.48 | 4.44 |
| | 10 | 3.64 | 1.47 | 5.04 | 10.15 | 3.48 |
| | 10 | 5.52 | 1.27 | 4.00 | 10.79 | 9.00 |

[a] Values are indicated as micrograms per gram.
No recorded value indicates values of less than 0.01 μg/g.

TABLE 6—*Approximate location of NMFS trawling stations where 14 gravid dogfish were captured, Oct.-Nov. 1975.*

| Station Number | Number of Fish | Longitude | Latitude | Water Depth, m |
|---|---|---|---|---|
| 5 | 1 | 40°34′ | 73°24′ | 15 |
| 16 | 1 | 40°53′ | 72°38′ | 35 |
| 22 | 5 | 40°53′ | 72°11′ | 35 |
| 213 | 1 | 40°44′ | 69°40′ | 44 |
| 282 | 2 | 42°05′ | 67°21′ | 42 |
| 298 | 1 | 41°02′ | 68°51′ | 71 |
| 405 | 2 | 44°01′ | 66°29′ | 101 |
| 429 | 1 | 42°36′ | 69°08′ | 201 |

young embryos, the lower values in the whole mature fetus, and the expected high concentration in the liver indicate a rather uniform body burden of DDT in the parental group and a transfer of both DDT and PCB from the maternal liver to the egg and to the liver of the mature fetus. The similarities in the residue magnitude probably reflect the behavioral characteristic of similarly aged dogfish to school together.

The biology of this shark helps to explain the existence of the residues. The dogfish liver is important as a storage body and may be as much as 75 percent lipid. In dogfish studies with $^{14}C$-labeled DDT, it was found that the pesticide passed readily through the intestinal wall, that about 85 percent of it was stored immediately in the liver, and that it was probably not metabolized further or excreted at any appreciable rate [*7*]. The dogfish is primarily a pelagic feeder, its diet consisting largely of herring and other plankton-feeding fish, and, on occasion, it feeds on plankton as well [*8*]. Consequently, its food would be subject to direct contamination with persistent chemicals resulting from atmospheric fallout. Finally, these sharks live 25 to 30 years and, more importantly, require 18 to 20 years to reach sexual maturity and produce their first eggs [*9*]. This suggests that the gravid females caught in 1975 spent the first decade of their lives between 1955 and 1965, a period when more than 220 000 kilos of DDT (active ingredient) were applied in just one coastal county in New Jersey [*10*]. About half of this amount was aerially applied, and both the prevailing winds and the water currents would have carried the volatilized or suspended DDT over the New England fishing grounds and the general area occupied by this population of dogfish.

We suggest that the parent stock of the juvenile sharks studied acquired a majority of its DDT residues in the period 1955–1965. The use of DDT for mosquito control was discontinued after 1966 in the New Jersey coastal marshes. The existing residues in dogfish appear to be the logical consequence of exposure to DDT, at its greatest biological availability, for perhaps ten years and to PCBs, with their relatively small biological availability in the ocean environment, for the past 20 years.

TABLE 7—*Residues of DDT and PCB in embryonic tissues excised from 14 gravid dogfish.*[a]

| Tissue | No. of Tissues | No. of Analyses | DDT | TDE | DDE | ΣDDT | PCB |
|---|---|---|---|---|---|---|---|
| Eggs | 6 | 3 | 0.27 | 0.12 | 0.27 | 0.66 ± 0.2 | 0.38 ± 0.07 |
| Fetus with yolk sac | 26 | 3 | 0.22 | 0.16 | 0.36 | 0.74 ± 0.4 | 0.50 ± 0.07 |
| Mature fetus without liver | 23 | 3 | 0.06 | 0.05 | 0.08 | 0.19 ± 0.08 | 0.10 ± 0.08 |
| Mature fetal liver | 33 | 4 | 0.63 | 0.42 | 0.94 | 1.99 ± 0.3 | 1.01 ± 0.3 |

[a] Values are indicated as micrograms per gram wet weight.

It would seem that this residue problem is not only species-specific but also restricted geographically; however, it is not unique. A similar situation has been described for dogfish in Spain [*11*]. The livers of dogfish collected in 1970 in the Mediterranean Sea had DDT residues as high as 18 $\mu g/g$ or about five times as much as those in dogfish collected on Spain's Atlantic coast. This pesticide has been used extensively in Mediterranean countries since the latter years of World War II for malaria-mosquito control.

The considerable variation in residue magnitude in different samples from juvenile dogfish indicates that there is significant attenuation of the compounds in their transmittal to successive broods of the same female. However, if a major part of the female's body burden of DDT and PCB is transmitted to the first brood, there is the distinct possibility that these residues will reappear in some segments of the juvenile dogfish population in 20-year cycles for some few shark generations to come.

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